

Mounding in Epitaxial Surface Growth

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For the past two decades the Villain-Lai-Das Sarma equation has served as the theoretical framework for conserved surface growth processes, such as molecular-beam epitaxy. However some phenomena, such as mounding, are yet to be fully understood. In the following, we present a systematic analysis of the full, original Villain-Lai-Das Sarma equation showing that mound forming terms, which should have been included initially on symmetry grounds, are generated under renormalisation. A number of widely studied Langevin equations are recovered as limits or trivial fixed points of the full theory.

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Crystal surface growth, in which the interface is driven by the deposition of new material from a process such as molecular-beam epitaxy (MBE), has been extensively studied [1–4]. Although the behaviour of surface growth in the absence of conservation generically belongs to the Kardar-Parisi-Zhang (KPZ) [5] universality class, frequently the growth condition for MBE allow the imposition of conservation laws that prohibit KPZ behaviour. Typically experimental observations of MBE on semiconductor wafers are compared with numerical simulations run on lattice models, popularly the Wolf-Villain (WV) [6] and the Das Sarma-Tamborenea (DT) [7] models, and continuum Langevin equations postulated from physical considerations. Analytically, epitaxial surface growth is almost exclusively modeled using the Villain-Lai-Das Sarma (VLDS) [8, 9] equation. Although successful, the theoretical framework seems somewhat incomplete. No clear picture has emerged over the alleged exactness of scaling relations and the mechanism of mound formation is unaccounted for. In the following we explain why mounding is, and ought to be, observed either transiently or stably in MBE/VLDS models; mound formation arises naturally from a term in the Langevin equation that has been variously missed, overlooked or discarded in the literature. A resolution to apparent disagreements and misunderstandings on the nature of the scaling relations and coupling renormalisation is offered. A complete theoretical picture for the behaviour and the different regimes of epitaxial surface growth as originally envisaged two decades ago is presented, with the full theory for conserved surface growth via ideal MBE given by:

$$\partial_t \phi(\mathbf{x}, t) = \nu_2 \nabla^2 \phi - \nu_4 \nabla^4 \phi + \lambda_{13} \nabla (\nabla \phi)^3 + \tilde{\lambda}_{22} \nabla^2 (\nabla \phi)^2 + \tilde{\kappa} \nabla (\nabla \phi \nabla^2 \phi) + \eta(x, t), \quad (1)$$

where the field $\phi(\mathbf{x}, t)$ is the surface (height) displacement at \mathbf{x} in the co-moving frame atop a d dimensional substrate at time t . Growth is subject to the white noise η with the usual correlator $\langle \eta(x, t) \eta(x', t') \rangle =$

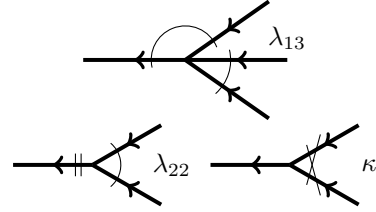


FIG. 1: The vertices of the three non-linearities in Eq. (1). Thick lines with arrows denote the bare propagator $(-\omega + \nu_2 \mathbf{k}^2 + \nu_4 (\mathbf{k}^2)^2)^{-1}$. Arcs and dashes drawn in narrow lines denote inner products of factors of \mathbf{k} . The vertex corresponding to κ has all factors of \mathbf{k} on one side, in the form $(\mathbf{k}_1 \times \mathbf{k}_2)^2 = \mathbf{k}_1^2 \mathbf{k}_2^2 - (\mathbf{k}_1 \cdot \mathbf{k}_2)^2$.

$2\Gamma^2 \delta^d(x - x') \delta(t - t')$. Ideal MBE [9] was proposed as “atomistic stochastic growth without any bulk defects or surface overhangs driven by atomic deposition in a chemical-bonding environment where surface relaxation can occur only through the breaking of bonds.” This constrains the surface dynamics to obey mass conservation; the deterministic evolution being cast as the divergence of some current, thus ruling out a KPZ term.

Re-writing two of the couplings in a computationally convenient form using $\tilde{\lambda}_{22} = \lambda_{22} - \kappa/2$ and $\tilde{\kappa} = \kappa$:

$$\underbrace{\left(\lambda_{22} - \frac{\kappa}{2}\right)}_{\tilde{\lambda}_{22}} \nabla^2 (\nabla \phi)^2 + \underbrace{\kappa}_{\tilde{\kappa}} \nabla (\nabla \phi \nabla^2 \phi) = \lambda_{22} \nabla^2 (\nabla \phi)^2 + \kappa \left[\nabla (\nabla \phi \nabla^2 \phi) - \frac{1}{2} \nabla^2 (\nabla \phi)^2 \right] \quad (2)$$

Giving rise to the vertices shown in Fig. 1. The original formulation of this Langevin equation [8, 9] describing epitaxial growth on a d dimensional substrate did not contain the κ term Eq. (2). However it should appear in two ways. Either it should be there from the start from the same symmetry and conservation arguments that produce λ_{22} . Or if it is absent it is generated any-

way from λ_{13} and λ_{22} under renormalisation of the full theory Eq. (1). It is this κ non-linearity that provides a natural mound forming mechanism. The κ notation has been used in keeping with the term's appearance in an unrelated restricted solid-on-solid model [10].

The original VLDS formulation was derived by considering the most general fourth order equation consistent with the symmetries of the problem, with further refinements coming from other physical insights. Later a second way of deriving the equation was sought by considering the transition rules on a lattice model of the surface [11–14], finding ways to turn them into a continuum equation and dropping terms considered to be irrelevant for the scaling behaviour (seeking to capture the “qualitative features of the surface morphology” [14]). Virtually all of the analysis of the continuum equations has been done using the Dynamic Renormalisation Group (DRG), a technique that is an extension of Wilson’s approach to Renormalisation Group (RG) calculations, in that a cutoff is used and momentum shells are integrated out to find the differential flow equations of the couplings (*e.g.* [1]). Instead of using DRG one can instead turn the Langevin equation in question into a field theory, the programme pursued below. This entails forming an action constrained by the Langevin equation in order to produce a (moment) generating functional. This approach was developed independently by Janssen [15] and De Dominicis [16].

Prosecuting a renormalised field theory of Eq. (1) to one loop various theories of epitaxial growth are recovered as fixed points or limiting cases. In all dimensions Eq. (1) displays Edwards-Wilkinson (EW) behaviour for ν_2 greater than the critical point ν_2^c , with $\nu_2^c = 0$ in the absence of non-linearities. For $\nu_2 < \nu_2^c$ the bare propagator acquires a pole at the characteristic wavelength $\sqrt{\nu_4/\nu_2}$. In dimensions above the critical dimension, $d_c = 4$, Eq. (1) is trivially governed by Mullins-Herring (MH) behaviour at critical $\nu_2 = \nu_2^c$. However, in dimensions $d = d_c - \epsilon < d_c$, the non-linearities λ_{13} , λ_{22} and κ are all (equally) relevant and produce the non-trivial scaling behaviour characterised below. In the presence of λ_{13} , the critical ν_2^c becomes negative and there is no longer a generic mechanism guaranteeing $\nu_2 = \nu_2^c$ as exists in the case $\nu_2^c = 0$, *i.e.* the non-trivial scaling is visible only after tuning to a critical point. This is the reason why λ_{13} has been dismissed originally. However, as pointed out by Haselwandter and Vvedensky [13], the non-trivial behaviour of the full theory might still be visible on an intermediate scale, beyond which EW might rule. In particular, given that κ is generated under renormalisation, finite mounding is expected to occur generically. What is normally referred to as the VLDS equation, however, is Eq. (1) with ν_2 , λ_{13} , κ set to zero.

The ultraviolet is regularised in a perturbation theory in small $\epsilon = d_c - d > 0$, where the reparamaterised,

dimensionless couplings read:

$$g = \frac{\Gamma^2}{(4\pi)^2} \frac{\lambda_{13}}{\nu_4^{2-\epsilon/2}} \quad \lambda = \frac{\lambda_{22}^2}{\nu_4 \lambda_{13}} \quad \chi = \frac{\kappa}{\lambda_{22}} \quad (3)$$

The renormalisation of the couplings is determined by accounting for all logarithmically divergent diagrams contributing to the proper vertices $\Gamma^{(1,1)}$, $\Gamma^{(1,2)}$ and $\Gamma^{(1,3)}$, the functional derivatives of the Legendre transform Γ of the cumulant generating function, *i.e.*:

$$\begin{aligned} \Gamma^{(n,m)}(\mathbf{k}_1, \dots, \omega_{n+m}; \nu_2, \nu_4, \Gamma^2, \lambda_{13}, \lambda_{22}, \kappa) = \\ \prod_{i=1}^n \frac{\delta}{\delta \tilde{\phi}(\mathbf{k}_i, \omega_i)} \prod_{j=1}^m \frac{\delta}{\delta \phi(\mathbf{k}_{n+j}, \omega_{n+j})} \\ \Gamma\left([\phi], [\tilde{\phi}]; \nu_2, \nu_4, \Gamma^2, \lambda_{13}, \lambda_{22}, \kappa\right), \end{aligned}$$

as derived from the one-particle irreducible, amputated diagrams contributing to the corresponding correlation function. They give rise to the renormalisation of the couplings α in the form $\alpha^R = Z_\alpha \alpha$. The infrared, on the other hand, is regularised by the mass $\nu_2 \neq 0$, with $\nu_2^R = Z_2 \nu_2 \mu^{-2}$, renormalisation point $\nu_2^R = 1$ and arbitrary inverse length μ .

The infrared stable fixed point is found as a root of the set of beta-functions, $\beta_a = d \ln \alpha / d \ln \mu|_{\text{bare}}$, where the derivative is to be taken for every coupling α at constant bare couplings:

$$\begin{aligned} \tilde{\beta}_g &= -\epsilon + \left(5 - (2 - \epsilon/2)\lambda \left[\frac{5}{2}\chi^2 - 3\chi - 1\right]\right) g \\ \tilde{\beta}_\lambda &= \left(4 - \lambda \left[\frac{5}{2}\chi^2 - 3\chi - 1\right]\right) g \\ \tilde{\beta}_\chi &= \left(\frac{1}{2} + \frac{1}{\chi}\right) g, \end{aligned}$$

where we define $\tilde{\beta}_\alpha = \beta_\alpha / \alpha$ for convenience. The Wilson gamma-functions are correspondingly defined as $\gamma_\alpha = d \ln Z_\alpha / d \ln \mu|_{\text{bare}}$:

$$\begin{aligned} \gamma_2 &= 3g, \quad \gamma_4 = \lambda \left(\frac{5}{2}\chi^2 - 3\chi - 1\right) g, \\ \gamma_g &= \left(5 - (2 - \epsilon/2)\lambda \left[\frac{5}{2}\chi^2 - 3\chi - 1\right]\right) g, \\ \gamma_\lambda &= \left(4 - \lambda \left[\frac{5}{2}\chi^2 - 3\chi - 1\right]\right) g, \quad \gamma_\chi = \left(\frac{1}{2} + \frac{1}{\chi}\right) g, \\ \gamma_{22} &= \frac{9}{2}g, \quad \gamma_\kappa = \left(5 + \frac{1}{\chi}\right) g, \quad \gamma_{13} = 5g \end{aligned}$$

As the noise does not renormalise at any order, $\gamma_\Gamma = 0$.

The scaling behaviour of the proper vertices,

$$\begin{aligned} \hat{\Gamma}^{(n,m)}(\mathbf{k}, \omega; \nu_2, \nu_4, \Gamma^2, \lambda_{13}, \lambda_{22}, \kappa) \\ = l^{-\frac{m}{2}(d-4) - \frac{\alpha}{2}(d+4) + d + 4 - \gamma_4(\frac{1}{2}(m-n) + 1)} \\ \hat{\Gamma}^{(n,m)}\left(\frac{\mathbf{k}}{l}, \frac{\omega}{l^{\gamma_4+4}}; \nu_2 l^{\gamma_2 - \gamma_4 - 2}, \nu_4, \Gamma^2, \lambda_{13} l^{-2\gamma_4 - \epsilon + \gamma_{13}}, \right. \\ \left. \lambda_{22} l^{\gamma_{22} - \frac{3\gamma_4}{2} - \frac{\epsilon}{2}}, \kappa l^{\gamma_\kappa - \frac{3\gamma_4}{2} - \frac{\epsilon}{2}}\right) \quad (4) \end{aligned}$$

determines the exponents natural to the field theory:

$$\begin{aligned} \hat{\Gamma}^{(n,m)}(\mathbf{k}, \omega; \nu_2, \nu_4, \Gamma^2, \lambda_{13}, \lambda_{22}, \kappa) \\ = l^{-\frac{m}{2}(d-4+\eta) - \frac{\alpha}{2}(d+4+\eta) + d + 4 + \delta} \\ \hat{\Gamma}^{(n,m)}\left(\frac{\mathbf{k}}{l}, \frac{\omega}{l^z}; \frac{\nu_2}{l^{1/\nu}}, \nu_4, \Gamma^2, \frac{\lambda_{13}}{l^{\pi_{13}}}, \frac{\lambda_{22}}{l^{\pi_{22}}}, \frac{\kappa}{l^{\pi_\kappa}}\right), \quad (5) \end{aligned}$$

Where the hat, $\hat{\cdot}$, indicates that the Dirac δ function from momentum conservation by translational invariance has been divided out, $\Gamma^{(n,m)} = \delta(\mathbf{k}_1 + \dots + \mathbf{k}_{n+m})\delta(\omega_1 + \dots + \omega_{n+m})\hat{\Gamma}^{(n,m)}$ eliminating one pair of arguments \mathbf{k} , ω . Normally, growth exponents (α , z and $\beta = \alpha/z$) characterise the approach of stationarity from a flat initial configuration and the finite size scaling of the roughness. In a field theory this is not particularly germane, so the exponents are equivalently (but see [17]) defined on the basis of the two point correlation function:

$$\begin{aligned} \hat{\mathbf{G}}^{20}(\mathbf{q}, \omega) = a|\mathbf{q}|^{-(d+z+2\alpha)} \mathcal{G}\left(\frac{\omega}{b|\mathbf{q}|^z}\right) = -\hat{\Gamma}^{20}(\mathbf{q}, \omega) \\ \times \left|\hat{\Gamma}^{11}(\mathbf{q}, \omega)\right|^{-2} = 2\Gamma^2 \left|\mathbf{q}\right|^{\gamma_4+4} \hat{\Gamma}^{11}\left(\frac{\mathbf{q}}{|\mathbf{q}|}, \frac{\omega}{b|\mathbf{q}|^{\gamma_4+4}}\right) \Big|^{-2} \end{aligned}$$

With suitable metric factor a and b and universal scaling function $\mathcal{G}(x)$. In the following, we therefore focus on the exponents

$$\delta = -\gamma_4, \quad \nu = \frac{1}{\gamma_4 + 2 - \gamma_2}, \quad z = \gamma_4 + 4, \quad \alpha = \frac{\epsilon + \gamma_4}{2}. \quad (6)$$

The simultaneous roots of the beta-functions give the fixed points of the theory. The infrared stable one at

$$\chi = -2, \quad \lambda = 0 \quad \text{and} \quad g = \epsilon/5 \quad (7)$$

gives $\gamma_4 = 0$ and $\gamma_2 = 3\epsilon/5$ and thus

$$\delta = 0, \quad \nu = \frac{1}{2} + \frac{3\epsilon}{20}, \quad z = 4, \quad \alpha = \frac{\epsilon}{2}, \quad (8)$$

are the exponents of the full VLDS equation Eq. (1) at the critical point $\nu_2 = \nu_2^c$. These are the same α and z exponents as for MH. With $\lambda = 0$ this implies both λ_{22} and κ are zero at the fixed point. The renormalisation of the full theory Eq. (1) is driven by λ_{13} . In agreement with previous results [9] with only λ_{22} we find that λ_{22} and κ do not renormalise themselves at one loop. The contributions from diagrams involving only these two couplings

neatly cancel at one loop but not at higher orders [18]. While κ and λ_{22} do not generate each other, they do mix under renormalisation.

Of the two unstable fixed points, the trivial one, $g = 0$, deserves further attention. As $g = 0$ implies $\lambda_{13} = 0$ which causes problems with the definition of λ , Eq. (3), it is not legitimate to naively read all the gamma-functions as zero and extract exact MH behaviour. The scaling behaviour of this trivial fixed point is normally referred to as the VLDS fixed point, observed either by taking the limit $\lambda_{13} \rightarrow 0$ or by removing it from the initial Langevin equation [9, 14], possible as λ_{13} is not generated by λ_{22} or κ . Keeping χ and replacing λ by

$$\psi = \frac{\Gamma^2}{(4\pi)^2} \frac{\lambda_{22}^2}{\nu_4^{3-\epsilon/2}} \quad (9)$$

gives $\tilde{\beta}_\psi = -\epsilon - (3 - \epsilon/2)[(5/2)\chi^2 - 3\chi - 1]\psi$ and

$$\delta = \frac{\epsilon}{3}, \quad \nu = \frac{1}{2} + \frac{\epsilon}{12}, \quad z = 4 - \frac{\epsilon}{3}, \quad \alpha = \frac{\epsilon}{3}, \quad (10)$$

where z and α are the traditional one loop VLDS exponents [9]. If only the λ_{22} non-linearity is present, that is $\chi = 0$ is taken at the trivial fixed point, then Eq. (1) becomes the original VLDS formulation. Interestingly the corrections to scaling at two-loops predicted by Janssen [18] on the basis of λ_{22} alone were found to be too small in numerical lattice simulations [19–21], this may be due to a κ correction. Renormalisation does however impose bounds on χ , or the fixed point of ψ would oblige an unphysical ν_4 . To one loop, in order to get sensible results $(5/2)\chi^2 - 3\chi - 1$ needs to be negative, $\chi \in [\frac{3-\sqrt{19}}{5}, \frac{3+\sqrt{19}}{5}]$, sufficiently large κ violates this. However the same exponents emerge [22] if implemented without consideration of this.

We observe that the κ coupling on its own is equivalent to a model proposed by Escudero [23] to reproduce VLDS behaviour. The non-linearity proposed in two dimensions, $\partial_{xx}\phi \partial_{yy}\phi - (\partial_{xy}\phi)^2$, is exactly the vertex parameterised by κ in Eq. (2). Pursuing the calculation with only κ is especially straight forward, power counting reveals that diagrams for its renormalisation constructed solely from the κ and the noise vertices are always ultra-violet finite; in the absence of any other coupling κ is not renormalised at any order. The only renormalisation is of the propagator, with one diagram at one-loop order. Hence scaling laws are not corrected to any order, yet Janssen's general insight is not wrong [18]; it is a peculiarity of having only κ that leads to non-renormalisation of the coupling, as opposed to a neat cancellation (only) at one-loop when λ_{22} is also present. Perhaps other analyses finding exact scaling laws have inadvertently examined this case rather than the VLDS equation. It is now quite apparent why this model reproduces VLDS exponents. It is now also apparent that the model's infrared stable fixed point is unphysical (to one loop), and its behaviour thus not assessable by perturbation theory.

As has been observed from its two-dimensional form [23], the κ non-linearity favours mound formation. In addition to Ehrlich-Schwoebel (ES) barriers expressed through ν_2 [3, 4] it provides a natural mechanism at the level of the continuum equations for mound formation. In the presence of λ_{13} , at the critical point, mounding is suppressed on the large scale, Eq. (7), yet visible at and below (transient) length scales $\propto \kappa^{5/(2\epsilon)}$, Eq. (4). Appealing to recent work done on coupling flow in models of ideal MBE [13] provides some theoretical justification for transient observation of mounding. At the trivial fixed point, on the other hand, VLDS scaling applies generically as ν_2^c does not suffer an additive renormalisation, and mounding may be present on all length scales.

A connection may also be made to the dynamics used in lattice models of ideal MBE. There have been several investigations into the link between rules of movement in lattice models to the terms in continuum equations that represent them, usually concentrating on one-dimension [24]. Hugston and Ketterl [25] showed that going from lattice rules that seem intuitive, or even computationally convenient, to continuum equations is subtle and fraught with unintended consequences. Step edge diffusion [26], appearing in two-dimensions, has been proffered as an additional mechanism to ES barriers that leads to unstable mounding, for example in the two-dimensional WV model [27]. The lattice rules for the DT model are slightly different and result in EW behaviour instead of mounding in two-dimensions. Differences in lattice rules may well be the distinction between having κ or not in the continuum equation for a lattice model.

Observing that the terms parameterised by λ_{22} and κ are equivalent in one-dimension offers an explanation as to why κ was absent in the original derivation of the VLDS equation. It was not generated by renormalisation since the analysis immediately focused on $\lambda_{13} = 0$ to prevent ν_2 generation. If ever $\lambda_{13} \neq 0$ then λ_{22} would be set to zero “without loss of generality”, as λ_{13} was deemed “more relevant” [28], further analysis halted by the generation of ν_2 . One analysis [13, 14] did not commit this omission, but unfortunately missed the diagram generating κ . A similar commentary applies to the use of master equations to generate continuum equations from lattice rules. The most telling sign is seen when, in developing a continuum equation for the WV model, derivatives are rearranged in one-dimension and the result carried over to higher dimensions where, however, it is invalid (e.g. Eq. (23) from Eq. (21) in [12]). One might wonder whether basic errors in multivariate calculus [29], such as assuming wrongly that $\nabla(\nabla\phi)^3$ equals $3(\nabla^2\phi)(\nabla\phi)^2$ [24] have thus far concealed the κ term. Fourier transforming such terms immediately reveals the problem [29].

In summary we have shown that the original VLDS formulation generates a mounding term, κ , overlooked in previous studies, partly due to non-linearities being deliberately omitted, partly the renormalisation schemes

employed missed the generation of κ and partly from an apparent misunderstanding of basic multivariate calculus. This κ term might effectively capture certain lattice rules that give rise to mounding in computer simulations; inadvertently studying models with only κ present may lead to concluding scaling relations are in general exact, whereas the presence of κ with λ_{22} may account for differences in expected scaling corrections.

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